

Thermal Conductivity of the $S = 1/2$ Quasi-One-Dimensional Ferromagnetic Spin System CsCuCl_3

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Abstract. We have measured the thermal conductivity along the c -axis, $\kappa_{\parallel c}$, parallel to ferromagnetic spin-chains of single crystals of the $S = 1/2$ quasi-one-dimensional spin system CsCuCl_3 in magnetic fields up to 14 T, in order to investigate the thermal conductivity due to spins, κ_{spin} , and the change of thermal conductivity corresponding to the change of the spin state. In the temperature dependence of $\kappa_{\parallel c}$, no contribution of κ_{spin} has been observed, while a dip has been observed at the antiferromagnetic phase transition temperature, T_N . Furthermore, it has been found that $\kappa_{\parallel c}$ at a low temperature of 5.1 K below T_N changes with increasing field perpendicular to the c -axis in good correspondence to the field-induced change of the spin state.

1. Introduction

Thermal conductivity in low-dimensional quantum spin systems has attracted interest, owing to the large contribution of the thermal conductivity due to spins, κ_{spin} [1, 2]. Especially, a number of studies on κ_{spin} have been carried out in the spin quantum number $S = 1/2$ one-dimensional (1D) antiferromagnetic (AFM) systems [3, 4, 5, 6, 7, 8, 9, 10, 11]. The large contribution of κ_{spin} has been observed as a peak or a shoulder in the temperature dependence of the thermal conductivity along spin chains together with the contribution of the thermal conductivity due to phonons, κ_{phonon} . In the 1D ferromagnetic (FM) systems, on the other hand, the contribution of κ_{spin} has not been observed clearly, though several studies on κ_{spin} have been performed [12, 13, 14, 15, 16].

The compound CsCuCl_3 belongs to the so-called ABX_3 family including well-known quasi-1D spin systems with frustration. The compound contains linear spin-chains of face-sharing CuCl_6 octahedra running along the c -axis. The spin chains separated by Cs^+ ions form an equilateral triangular lattice in the c -plane. The intrachain exchange interaction J is FM and estimated as ~ -28 K, while the interchain exchange interaction J' is AFM and estimated as ~ 5 K [17]. Therefore, frustration is expected to exist more or less between the spin chains. The CuCl_6 octahedra are distorted due to the Jahn-Teller effect, so that the Dzyaloshinsky-Moriya interaction is induced between spins along the c -axis. An AF long-range order appears at the phase transition temperature, T_N , ~ 10.7 K, owing to J' [18]. The spin structure at low temperatures below T_N is helical with the pitch of $\sim 5.1^\circ$ along the c -axis owing to the

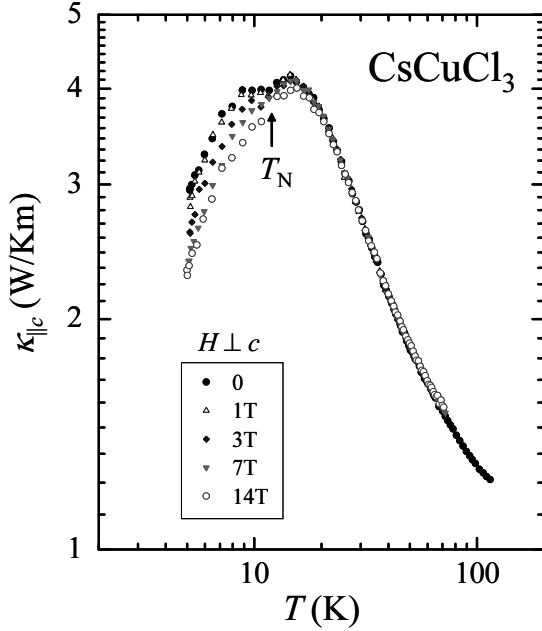


Figure 1. Temperature dependence of the thermal conductivity along the c -axis, $\kappa_{||c}$, parallel to spin chains in zero field and magnetic fields perpendicular to the c -axis.

combination of J and the Dzyaloshinsky-Moriya interaction. In the c -plane, the so-called 120° spin structure is formed owing to J' and the spin directions are confined in the c -plane [18]. Moreover, it has been known that the pitch decreases by the application of magnetic field perpendicular to spin chains [19, 20, 21].

In this paper, we have measured the thermal conductivity along the c -axis, $\kappa_{||c}$, parallel to spin chains of CsCuCl_3 in order to investigate the contribution of κ_{spin} . Furthermore, we have also investigated the relation between the spin state and the behavior of the thermal conductivity, because thermal conductivity is a good probe detecting a change of the spin state in a spin system via the change of the scattering rate of heat carries [22, 23, 24, 25, 26].

2. Experimental

Single crystals of CsCuCl_3 were grown from solution. Thermal conductivity measurements were carried out by the conventional steady-state method in magnetic fields up to 14 T.

3. Results and Discussion

Figure 1 shows the temperature dependence of $\kappa_{||c}$ parallel to spin chains in zero field and magnetic fields perpendicular to the c -axis at low temperatures below ~ 100 K. It has been found that $\kappa_{||c}$ show a peak around 10 K with a dip at T_N in zero field. This kind of dip at T_N has been observed in several antiferromagnets [5, 27, 28], and is interpreted as being due to the strong scattering of heat carries caused by the critical fluctuations around T_N or due to the increase of the mean free path of heat carries just below T_N on account of the marked reduction of the scattering caused by the development of the long-range order. Therefore, this behavior of $\kappa_{||c}$ in zero field is likely to be mainly due to the contribution of κ_{phonon} affected by the scattering of phonons by magnetic excitations. The contribution of κ_{spin} cannot be observed clearly in CsCuCl_3 . Recently, single crystals of CsCuCl_3 of a single domain with homochirality of spins have been grown by Kousaka *et al.* [29]. On the other hand, our single crystals of the zigzag shape may be of multi-domains. Therefore, κ_{spin} might strongly be suppressed due to the strong scattering of magnons at the domain boundaries.

By the application of magnetic field perpendicular to the c -axis, it has been found that $\kappa_{||c}$ at

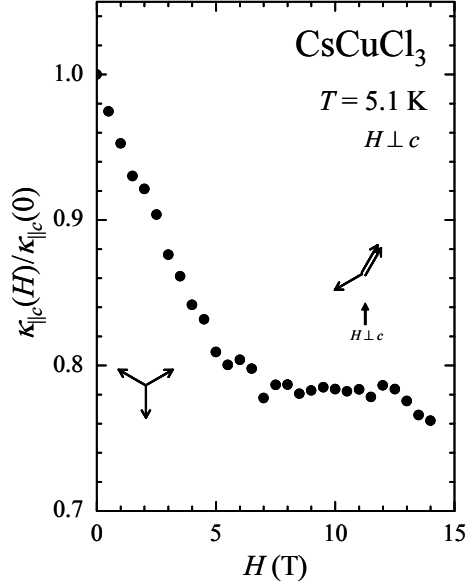


Figure 2. Magnetic-field dependence of the thermal conductivity along the c -axis normalized by the value in zero field, $\kappa_{||c}(H)/\kappa_{||c}(0)$, of CsCuCl_3 in magnetic fields perpendicular to the c -axis. Three arrows on the left and right sides mean 120° and coplanar 2-1-type spin structures, respectively.

low temperatures below T_N is suppressed, as shown Fig. 1. To see the suppression in detail, the magnetic-field dependence of $\kappa_{||c}$ normalized by the value in zero field, $\kappa_{||c}(H)/\kappa_{||c}(0)$, at 5.1 K is shown in Fig. 2. It has been found that $\kappa_{||c}(H)/\kappa_{||c}(0)$ monotonically decreases with increasing field up to ~ 9 T, becomes constant between ~ 9 T and ~ 12 T, and then decreases again above ~ 12 T. This behavior is very similar to the magnetic-field dependence of the incommensurability, δ , which is proportional to the pitch of the helical spin structure along the c -axis [19, 20, 21]. It is known that the helical and 120° spin structures are maintained in low magnetic fields below ~ 9 T, though the pitch of the helical structure along the c -axis decreases with increasing field [21]. The 120° spin structure in the c -axis is expected to become unstable by the application of magnetic field in the c -plane. Therefore, it is likely that $\kappa_{||c}$ is suppressed with increasing field due to the enhancement of the magnetic fluctuations scattering phonons.

In a region between 9 T and 16 T, it is known that the helical and the so-called coplanar 2-1-type spin structures, where two thirds of spins are roughly parallel and the others are roughly antiparallel to the magnetic-field direction as shown in Fig.2, are formed. The constant value of $\kappa_{||c}(H)/\kappa_{||c}(0)$ in this region implies that the number of magnetic excitations scattering phonons does not change so much. This is consistent with the locking phenomenon of the spin structure in this region observed in the neutron scattering experiment [21] and with the plateau-like behavior in this region observed in the magnetization curve also [30].

In high magnetic fields above ~ 12 T, on the other hand, the suppression of $\kappa_{||c}$ can be explained by the enhancement of magnetic fluctuations scattering phonons, because the field-induced transition to the non-helical commensurate phase with $\delta = 0$ takes place at ~ 16 T [20, 21]. The behavior of $\kappa_{||c}$ is also roughly consistent with the neutron scattering result that the value of δ starts to decrease at ~ 14 T with increasing field [20, 21].

4. Conclusions

We have measured $\kappa_{||c}$ parallel to spin chains of CsCuCl_3 , in order to investigate the presence or absence of κ_{spin} and the change of the spin state. Neither peak nor shoulder due to the contribution of κ_{spin} has been observed in the temperature dependence of $\kappa_{||c}$ in zero field. This may be due to the strong scattering of magnons at boundaries of domains with homochirality of spins in the helical spin structure along the c -axis. By the application of magnetic field

perpendicular to the c -axis up to 14 T, it has been found that $\kappa_{\parallel c}$ at 5.1 K below T_N decreases with increasing field up to ~ 9 T, is constant between ~ 9 T and ~ 12 T, and decreases again above ~ 12 T. These behaviors have been explained as being due to the change of κ_{phonon} caused by the change of the scattering of phonons by magnetic excitations due to the field-induced change of the spin state.

Acknowledgments

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